

# Turning Carbon Directly into Electricity

*The carbon conversion fuel cell pushes the efficiency of using fossil fuels to generate electricity closer to theoretical limits.*

**T**HE long-term energy—and environmental—future of the United States is much in the headlines these days. Helping to make the prospects brighter is a team of Lawrence Livermore scientists working to develop a method for producing electricity that is safe, relatively simple, remarkably efficient, and kind to the environment.

Called direct carbon conversion, the process has been demonstrated convincingly in the laboratory over the past year. The electrochemical process converts carbon particles, obtained from different fossil fuels, directly into

electricity without the need for such traditional equipment as steam-reforming reactors, boilers, and turbines.

The breakthrough Lawrence Livermore method, the result of a two-year study funded by the Laboratory Directed Research and Development Program, pushes the efficiency of using fossil fuels for generating electricity far closer to theoretical limits than ever before. If adopted on a large scale, direct carbon conversion would help to conserve precious fossil resources by allowing more power to be harnessed from the same amount of fuel. It would

also improve the environment by substantially decreasing the amount of pollutants emitted into the atmosphere per kilowatt-hour of electrical energy that is generated. Perhaps most important, it would decrease emissions of carbon dioxide, which are largely responsible for global warming.

“What if we could nearly double the energy conversion efficiency of fossil fuels in electric power generation over the conversion efficiency of today’s coal-fired power plants—which is about 40 percent—and thereby cut the carbon dioxide emissions per kilowatt almost in half?” asks lead researcher John Cooper, scientific capability leader for electrochemistry and corrosion in Lawrence Livermore’s Chemistry and Materials Science Directorate. “And what if we could produce a pure carbon dioxide byproduct for sequestration or industrial use at no additional cost of separation while avoiding the air pollution problems associated with combustion?”

Cooper explains that direct carbon conversion requires a unique kind of fuel cell. A fuel cell is an electrochemical device that efficiently converts a fuel’s chemical energy directly to electrical energy without burning the fuel. However, instead of using gaseous fuels, as is typically done, the new technology uses aggregates of extremely fine (10- to 1,000-nanometer-diameter) carbon particles distributed in a mixture of molten lithium, sodium, or potassium carbonate at a temperature of 750 to 850°C. The overall cell reaction is carbon and oxygen (from ambient air) forming carbon dioxide and electricity.

The reaction yields 80 percent of the carbon-oxygen combustion energy as electricity. It provides up to 1 kilowatt of power per square meter of cell surface area—a rate sufficiently high for practical applications. Yet no burning of the carbon takes place.

### No Water to Boil

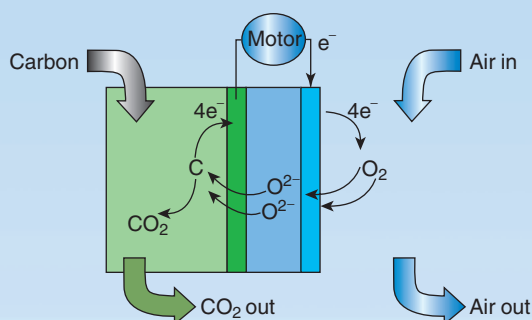
“We’re not burning fossil fuels to boil water to drive turbines and dynamos to generate electricity,” says Lawrence Livermore electrochemist Nerine Cherepy, who has been researching the breakthrough concept. “This is a simpler, more efficient, and more environmentally friendly process that obtains the greatest possible fraction of energy from the starting fossil fuel with little waste heat.”

The thermodynamic efficiency of the direct carbon conversion cell exceeds the 70-percent requirement of the next-generation fuel cell envisioned by the Department of Energy. In contrast, conventional coal- and natural-gas-fired power plants are typically between 35- and 40-percent efficient. Combined-cycle pilot plants that burn natural gas in multistage turbines now operate at 57-percent efficiency, based on the higher heating value of the fuel. (Higher heating value, or HHV, is the total amount of heat released when a fuel is burned completely and the products are returned to their natural, room-temperature states.) High-temperature fuel cell hybrid systems (fuel cells combined with turbines), such as a technology developed by Westinghouse, are expected to operate on natural gas at 60-percent HHV.

Direct carbon conversion can use fuel derived from many different sources, including coal, lignite, petroleum, natural gas, and even biomass (peat, rice hulls, corn husks). Cooper notes that 90 percent of Earth’s electric energy comes from the burning of fossil fuels. Half of our fossil-fuel resources is coal, and 80 percent of the coal belongs to the United States and Canada, the former Soviet Union, and China. Coal-fired plants produce 55 percent of U.S. electricity—as well as large amounts of pollutants. As a result, the vast energy reserves of coal remain underused. Direct carbon conversion has the potential to be the long-sought “clean coal” technology.

The carbon-air fuel cell gives off a pure stream of carbon dioxide that can be captured without incurring additional costs of collection and separation from smokestack exhausts. The stream of carbon dioxide, already only a fraction of current processes, can be sequestered or used for oil and gas recovery through existing pipelines. (Lawrence Livermore environmental scientists are studying the sequestering of carbon dioxide in geologic formations as part of a Department of Energy effort. See *S&TR*, December 2000, pp. 21–23.)

Pyrolysis—the thermal decomposition method used to turn hydrocarbons into hydrogen and tiny pure carbon particles



Carbon (C) and oxygen ( $O_2$ ) can react in a high-temperature fuel cell with the carbon, delivering electrons ( $e^-$ ) to an external circuit that can power a motor. The net electrochemical reaction—carbon and oxygen forming carbon dioxide—is the same as the chemical reaction for carbon combustion, but it allows greater efficiency for electricity production. The pure carbon dioxide ( $CO_2$ ) product can be sequestered in an underground reservoir or used to displace underground deposits of oil and gas.

used in direct carbon conversion—consumes less energy and requires less capital than the electrolysis or steam-reforming processes required to produce hydrogen-rich fuels. Pyrolysis produces billions of kilograms of carbon blacks annually in the U.S. Carbon black is a disordered form of carbon produced by thermal or oxidative decomposition of hydrocarbons and is used to manufacture many different products, including tires, inks, and plastic fillers.

### Old Dream

Electricity direct from coal is one of the earliest dreams of electrochemical science. The first attempts date from the late 19th century, when Boston entrepreneur William Jacques fashioned a coal fuel battery that used coke electrodes in a molten sodium hydroxide electrolyte. Because the molten electrolyte became exhausted, Jacques's invention operated as an exhaustible battery, not as a fuel cell, despite impressive demonstrations on the kilowatt scale. Other problems included a buildup of ash entrained with the fuel, the cost of making the carbon anodes, and the difficulty of distributing carbon fuel electrodes to

the many cells. Efforts to develop practical carbon-based fuel cells during the 20th century, such as those tested at the Stanford Research Institute in the 1980s, were also hindered by the buildup of ash and by the costs and difficulties of carbon electrode manufacture.

The Lawrence Livermore approach circumvents the historic barriers to a coal fuel cell by using extremely fine, virtually ash-free, "turbostratic" carbon particles that contain small amounts of ash and have a high degree of structural disorder on the nanometer scale. The team found that turbostratic carbon particles, when mixed with molten carbonate to form a slurry, operate like rigid electrodes when the melt is brought into contact with an inert metallic screen. Exactly how the carbon particle delivers energy to the screen is under investigation, but reactive chemicals in the melt produced by the carbon are likely intermediates.

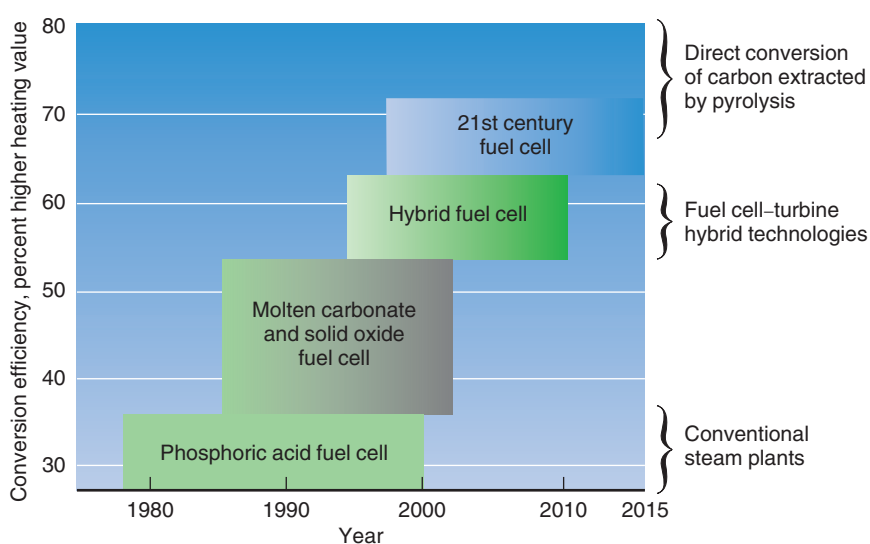
Also, the team found that carbon particles can be distributed pneumatically to individual cells by a small amount of carbon dioxide fed back to the cell from the continuously

produced carbon dioxide stream. (The pneumatic transport of carbon particles through complex equipment is a widespread industrial practice.)

The carbon particles and oxygen (ambient air) are introduced as fuel and oxidizer, respectively. The slurry formed by mixing carbon particles with molten carbonate constitutes the anode. The anode reaction is carbon and carbonate ions forming carbon dioxide and electrons. At the cathode, which is similar to that used in other high-temperature fuel cells, oxygen, carbon dioxide, and electrons from the anode form carbonate ions. A porous ceramic separator holds the melt in place and allows the carbonate ions to migrate between the two compartments.

The technology has been demonstrated in a number of small, experimental cells with reaction areas of about 3 to 60 square centimeters. The cells feature different designs and different materials, including stainless steel, ceramic, and sometimes graphite. Each cell type features tubes for gases to enter and exit the cell, thermocouples (for measuring temperature), and a reference electrode. Temperature is maintained by a

Various kinds of fuel cells using phosphoric acid, molten carbonate, and solid oxide yield electricity from methane fuel at efficiencies in the range of 35 to 55 percent of higher heating value. Using waste heat from the fuel cell in turbines (hybrids) can increase the total efficiency even further. The thermodynamic efficiency of the direct carbon conversion cell already exceeds the 70-percent efficiency goal of the 21st century fuel cell envisioned by the Department of Energy.





computer-controlled furnace. The computer also acquires continuous data on current and voltage.

In repeated tests, the cells deliver up to 0.1 watt continuously per square centimeter and are 80-percent efficient at 80 milliwatts per centimeter. Recently, using a new cell design that automatically regulates the amount of molten salt, the team has operated cells for days, simply by adding more carbon fuel.

### Doubly Attractive

The carbon–oxygen reaction is attractive in two unique ways, says Cooper. First, almost no entropy change occurs in the overall cell reaction. (Entropy is a measure of the disorder in a system. A significant entropy decrease would mean that the cell produces a great deal of waste heat.) Because the entropy change is close to zero for the carbon–oxygen reaction, 100 percent of the heat energy of combustion of the carbon can instead be converted by the cell into electrical energy under ideal conditions.

Second, the driving force for energy production, called electromotive force or maximum voltage, does not degrade

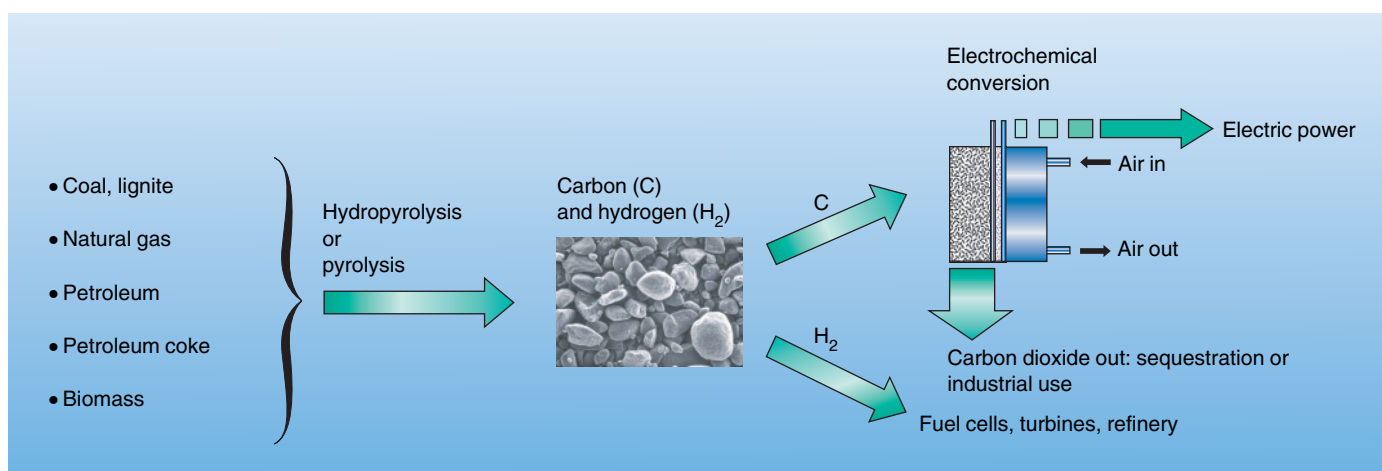
as the carbon is progressively consumed to make power and carbon dioxide, so the voltage remains constant. That means that in making a single pass through the cell, all the carbon is consumed at a maximum yet constant voltage.

“Realistically, we can get out a maximum of about 80- to 85-percent efficiency, based on the heating value of the carbon, when the cell is operated at a practical rate, which is about 100 milliamperes per square centimeter,” says Cooper. “The losses are primarily those associated with the sluggishness of electrode reactions and the electrical resistance of the cell. It was the two thermodynamic properties—zero entropy change and constant electromotive force—that first drew our attention to carbon as an attractive electrochemical fuel.” In contrast, the entropy decrease for the hydrogen–oxygen reaction in high-temperature fuel cells limits conversion efficiency to 70 percent of the fuel’s HHV, while electrical efficiencies (about 80 percent) and practical fuel use (about 80 percent) further reduce the total efficiency to below 50 percent. (See the [box on p. 10.](#))

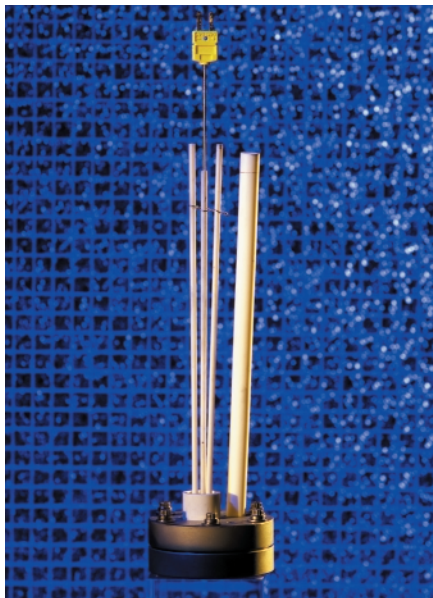
The part of a fuel’s combustion energy that is not converted to electric power appears as heat. Some of this heat could be used to generate steam and drive a turbine generator, as in hybrid systems. But the additional cost and complexity must be weighed against the comparatively small additional savings in fuel.

### Nanostructure Is Important

Cherepy and senior scientific associate Roger Krueger have tested a number of pure carbons that differ principally in the degree and nature of disorder on the nanoscale. They have correlated significant differences in the carbon fuels’ three-dimensional atomic structures with their electrochemical reactivities. The more disordered the carbon atoms, the more easily they yield electrons. Cherepy and Krueger are paying particular attention to turbostratic carbons, which feature planes of atoms arranged at different angles and with lots of defects at the edges that make the atoms more accessible for chemical reactions. (Graphite, in contrast, has a more ordered structure and is less reactive by a factor of about 1,000.)



In principle, any fossil fuel or biomass can be converted to electric power using direct carbon conversion. For natural gas and oil, pyrolysis (thermal decomposition) yields hydrogen and carbon. For dirtier resources (coals, biomass), the carbon may have to be extracted by reaction with hydrogen, followed by pyrolysis.



Simple cells in the laboratory are used to react carbon and atmospheric oxygen. These cells consist of a metal anode current collector, a ceramic matrix for holding the melt, and a metal screen for reacting the oxygen from the air.

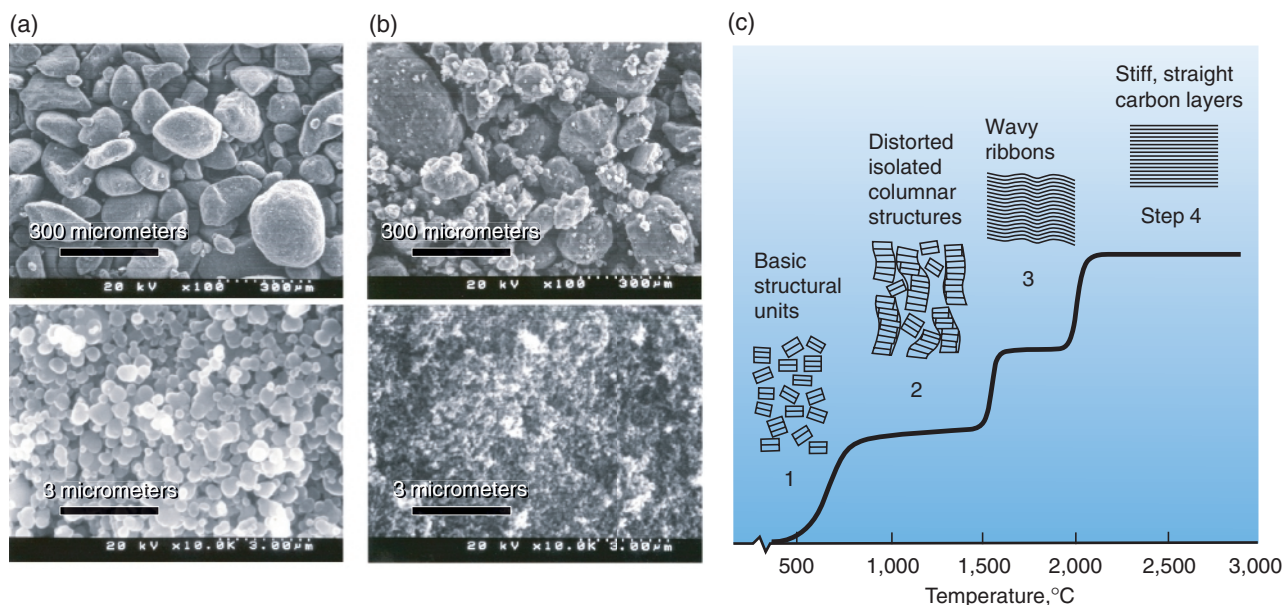
Those candidate turbostratic carbons exhibiting discharge rates of more than 20 milliamperes per square centimeter at 0.8 volts have been analyzed in greater detail with transmission electron microscopy and x-ray diffraction. Researchers in Livermore's Chemistry and Materials Science Directorate conducted some of these characterization tests, and other characterizations have been done by Kim Kinoshita and coworkers at Lawrence Berkeley National Laboratory.

Cherepy's investigation has focused largely on carbon blacks because they have the highest electrochemical reactivity of any carbon fuel yet tested. Made from a variety of sources, carbon blacks are the basis of a large commercial industry. Four and a half billion kilograms per year of carbon black (all turbostratic to various degrees) are produced annually for automobile tires, pigments, plastics fillers, wire insulation, and other

products. Although most carbon blacks contain about 0.02 to 0.05 percent residual ash, it should have no effect on system performance, cost, or cell lifespan because the rate of ash accumulation would be slow. (Carbon with 0.02-percent ash would clog the cell after about 50 years, five times the life expectancy of cell hardware.)

Among carbon blacks, a range of reactivities has been measured. For example, one carbon material had a peak power density of about 8 milliwatts per square centimeter while a second material measured almost 50 milliwatts per square centimeter. A third, the best material tested, yields energy at about 100 milliwatts per square centimeter and 100 milliamperes at 0.8 volts, sufficient for many fuel cell or battery applications.

Significant differences in microstructure and nanostructure were found in electron micrographs of the three samples, although all are nearly



The structure of the carbon material is the key to widely different electrochemical reactivities. The two carbon blacks in (a) and (b), revealed in photomicrographs at two magnifications, were produced by pyrolysis at different temperatures and started out as different fossil fuels. In (c), the degree of disorder of the carbon increases as the temperature of formation decreases from 2,000°C down to about 700 to 1,000°C.

pure carbon and look like black dusts. X-ray diffraction measurements showed that all had much greater spacing between layers of carbon atoms than does graphite. The x-ray data also revealed only small areas of crystallinity compared to graphite. Finally, the more reactive carbons have higher surface area and were found to oxidize more rapidly when exposed to high temperatures in air.

Cooper notes that the team is working to achieve a better understanding of the relationship between the nanostructure of carbons and their electrochemical reactivity in molten salts. A related goal is being able to predict carbon nanostructure from the conditions of pyrolysis and the nature of the starting materials undergoing pyrolysis. "Success here is critical to the economic attractiveness of the process and its ability to draw upon any fossil fuel resource," he says.

### One Class of Fuels, Many Sources

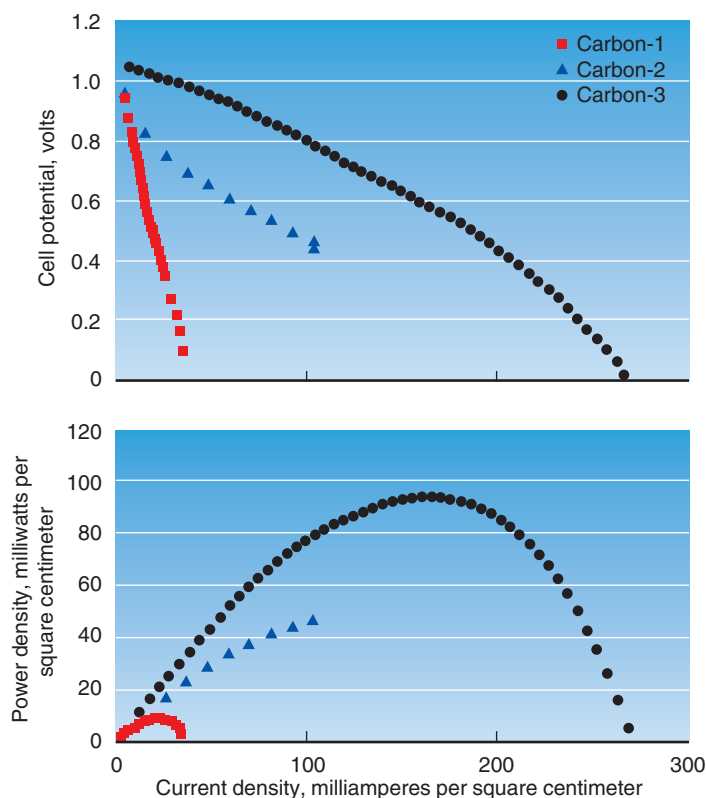
A significant advantage of direct carbon conversion is that practically any fossil fuel, including coal, lignite, biomass, natural gas, and petroleum, can produce turbostratic carbons. One method, pyrolysis, uses moderate temperatures (800 to 1,200°C) to produce a stream of elemental carbon particles and a stream of hydrogen gas from a pure hydrocarbon. The byproduct hydrogen gas can be sold for a number of uses, including chemical synthesis, combustion, and powering fuel cells. The pyrolysis step consumes 5 to 10 percent of the starting fuel value (1 to 2 percent is lost because of process inefficiencies). Some fossil fuels, such as coal and biomass, first require treatment with hydrogen under high pressure to produce a hydrocarbon that can then be pyrolyzed into carbon fuel and recyclable hydrogen. This treatment is called hydropyrolysis and has many variants.

One of the most intriguing options is using coal as a carbon source because of the nation's (and the world's) vast resources of coal and the difficulty in using coal as a clean energy source. Because of most coal's high sulfur and ash content, it must undergo hydropyrolysis or some other means of purification.

Turbostratic carbon from petroleum coke could be highly advantageous for the carbon conversion cell, says Cooper, because it would likely be the least expensive source of carbon fuel. Some 2 to 8 percent of all petroleum refining

ends up as petroleum coke, an inexpensive waste product that is naturally turbostratic and could be modified and used for direct carbon conversion. The amount of coke produced will increase as lighter crude resources become exhausted. Because coke commonly contains 0.25- to 5-percent sulfur, direct carbon conversion cells would require either coke refining or the use of graphite conductors in the carbon-air cell to prevent sulfur corrosion.

For natural gas, Cooper envisions small (100-kilowatt), transportable



Three carbon blacks are equally pure, are made by pyrolysis, and cost about the same, but they differ significantly in structure on the nanometer scale. As a result, their electrochemical reactivities are quite different. The two graphs depict the three carbons' voltage and power—two different functions of electrical current—as tested in a direct carbon conversion cell. Power densities (bottom) show carbon-3 reacting at a rate 10 times greater than carbon-1, providing about 100 milliwatts per square centimeter at 850°C. (Graphite, by comparison, is about 1,000 times less reactive than carbon-3.)

power stations that could be run from any natural gas pipeline. Such small power stations would be ideal in natural gas production fields; when a field becomes exhausted, the cell would be moved to a new location. Natural gas would be filtered and pyrolyzed at the wellhead. The resulting turbostratic carbon would go immediately to a direct carbon conversion cell, the hydrogen to a fuel cell, and hot carbon dioxide from the carbon cell used to displace more natural gas.

Direct carbon conversion might also make use of a significantly underused family of fuels that includes biomass, lignite, peat, and others. Some of this material, such as rice hulls, straw, and

corn stalks, is simply burned in the field after harvest. Antipollution regulations are increasingly making such burning unlawful. Instead, such material could be charred, and the carbon component extracted with hydropyrolysis.

In analyzing the various fuel options, the team, together with Meyer Steinberg from Brookhaven National Laboratory, has calculated the total HHV efficiencies for electric power generation through five different routes to the production of turbostratic carbons, including petroleum coke, refinery products, natural gas, and lignite coal. The findings were 80 percent for direct petroleum coke, 67 to 75 percent for natural gas

(methane), 72 percent for heavy oil, and 68 percent for lignite.

### Costs Keep Things Interesting

An important aspect of the research effort is estimating costs for electrical production and for cell components. Petroleum coke is by far the least expensive source of fuel (costing as little as 5 cent per kilogram) because it is the byproduct of the oil refining industry. In the carbon black industry, the pyrolysis step costs about 20 cents per kilogram of carbon produced and thus would contribute about 3 cents to the cost per kilowatt-hour of electricity generated using carbon-black fuel.

At this time, cost estimates are difficult to make. A final design for the

## Comparing Fuel Cells

Fuel cells use hydrogen, simple hydrocarbons such as methane, or carbon to produce electricity electrochemically rather than by burning them as fuels. Electrochemical means of providing electricity are generally much more efficient than burning fossil fuels in power plants to drive boilers and dynamos. The theoretical efficiencies of hydrogen or methane fuel cells top out at 69 percent and 90 percent, respectively, compared to 40-percent efficiency for typical power plants. The carbon-oxygen reaction that drives a direct carbon conversion fuel cell is unique: theoretically, all the potential combustion heat can be converted to electric power.

Methane and hydrogen fuel cells have other disadvantages. For one, the fuels are continuously diluted by their own reaction products as they are consumed. The voltage drops to ever-lower values, and as a result, not all of the fuel can be consumed. For carbon, no such dilution occurs, and all of the

incoming fuel can be used to make electricity at about the same rate and voltage. Hydrogen, methane, and carbon fuel cells have practical voltage efficiency, that is, they operate at 80 percent of the maximum cell voltage.

The total electrical efficiency of a fuel cell is the product of three factors: theoretical efficiency, the fraction of fuel used, and the voltage efficiency. Carbon has a high total efficiency because of the favorable thermodynamics of the carbon-oxygen reaction. The actual efficiencies of the hydrogen and methane cells achieved in pilot plants are listed in the table below.

Of course, different kinds and amounts of energy are used in making these fuels. Methane needs only to be extracted from natural gas—a low-cost technology. Hydrogen can be produced from nuclear and renewable energies without any production of carbon dioxide. Carbon can be derived at a low energy cost from nearly any fossil fuel.

**Comparison of efficiencies of fuel cells**

| Fuel     | Theoretical limit | Fraction of fuel used in practical operation | Fraction of voltage available at practical rate | Total efficiency (higher heating value) |
|----------|-------------------|--|---|---|
| Carbon   | 1.01              | 1.0  | 0.8   | 0.80                                    |
| Hydrogen | 0.69              | 0.75 to 0.85                                 | 0.8   | 0.41 to 0.47                            |
| Methane  | 0.90              | 0.75 to 0.85                                 | 0.8   | 0.54 to 0.61                            |

(Operating temperature of 750°C. Energy cost of fuel synthesis is excluded.)



hardware has not been settled on, and increases in power density are expected that inversely affect hardware size and cost. Nevertheless, the cost of the most expensive part of the cell—the commercial ceramic matrix holding the electrolyte and electrodes—is about \$200 per square meter (that is, about \$200 per kilowatt at 1 kilowatt per square meter). By comparison, modern gas turbine plants generate power at about \$350 per kilowatt. Currently, the cost of cell hardware is low enough to be interesting.

The sheer simplicity of the cell contributes to keeping costs down. The cell's fundamental thermodynamic properties mean almost no waste heat and full fuel consumption. Also,

because the carbon conversion process produces pure carbon dioxide ready for sequestration or industrial use, cell design does not need costly components to collect and scrub the carbon dioxide before storage or use.

Finally, cell components and fuel are nontoxic and relatively hazard-free. In particular, because the carbon–molten salt slurry does not explode if inadvertently brought into contact with air, no explosion-prevention safeguards need to be engineered into the cells.

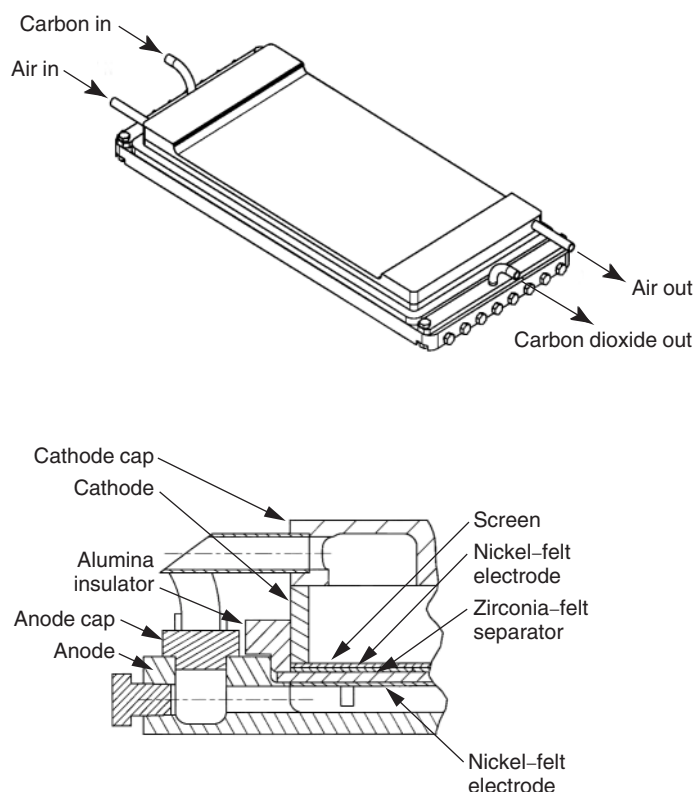
### Destined for the Grid

Cooper is thinking ahead to the day when direct carbon conversion units could be used to generate electric power for the grid. Such a power plant

would be “large, but not gigantic.”

A 3-gigawatt direct-carbon-conversion power plant, big enough to continuously supply some 3 million homes with about 1 kilowatt each, would only be the size of a large, two-story office building.

To achieve commercial adoption, however, requires greater understanding of the underlying science, especially the three-way relationship between conditions of pyrolysis, the resulting carbon nanostructure, and the electrochemical reactivity. While pyrolysis of natural gas and oil products to make turbostratic carbons is well known and widely practiced, the extraction of carbon from coal is less developed. “The extraction of carbon from coal, for example, by



An advanced cell design scales up the dimensions of tested cells to the 1,000-square-centimeter level. A maximum of 100 watts is expected from this design.



Nerine Cherepy and John Cooper assemble an experimental carbon conversion fuel cell.



## Key Points to Understanding Carbon Conversion and Its Potential

- No single solution exists to meet 21st century energy and environmental needs. Electrification of highway vehicles, conservation, advanced turbines, electrochemical conversion of fuels (as with direct carbon conversion), nuclear power, and renewable energy are all likely to be important.
- It is critically important to develop technologies that generate electric power much closer to theoretical limits—the best large-scale commercial technologies are only halfway there.
- Direct carbon conversion generates electricity from reacting carbon and oxygen in a fuel cell and makes a pure carbon dioxide product available for industrial use or sequestration.
- Using fossil energy as carbon in a carbon fuel cell produces little waste heat and consumes all the fuel in a single pass, thereby bringing total efficiencies of 70 to 80 percent into reach.

hydropyrolysis, needs to be developed if this approach is to aid the conversion of 50 percent of Earth's fossil fuels," says Cooper.

The team is planning to scale up a demonstration unit from the 3-watt experimental cell to a stackable, 100-watt engineering module with 1,000 square centimeters of active area. The large-scale experiments should reveal any materials and operational problems on a practical scale, especially during extended tests.

Meanwhile, the team is testing more carbon blacks from commercial suppliers and turbostratic carbon fuels from new sources, such as petroleum cokes and coals. The tests with coal will be particularly important because of its large-scale reserves.

Cooper points to the complex task of providing energy while controlling greenhouse gases, particularly carbon dioxide. "The solution is beyond the scope of power production technology alone," he says, noting that electrical

energy production currently accounts for just one-sixth of the total output of carbon dioxide. "Advanced combustion, fuel cells, nuclear and renewable energy, and conservation may all combine to help the situation

in a way that cannot presently be predicted."

The Livermore team considers it vitally important to develop a simple fuel cell technology that greatly increases the yield of electric energy from each unit of fossil fuel, uses fuels derived efficiently from almost any fossil fuel, significantly decreases the carbon dioxide released into the atmosphere, and makes it easy to capture the carbon dioxide for sequestration or other use.

Clearly, we're just beginning to hear about direct carbon conversion.

—Arnie Heller

**Key Words:** biomass, carbon black, carbon dioxide, coal, direct carbon conversion, fuel cell, global warming, hydrogen fuel cell, hydropyrolysis, natural gas, petroleum coke, pyrolysis, turbostratic carbon.

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## About the Scientist



**JOHN COOPER** received his B.A. in chemistry from Pomona College in 1968 and his Ph.D. in chemistry from the University of California at Berkeley in 1975. For 25 years, he has specialized in electrochemical science and engineering, with particular emphasis on fuel cells, fuel batteries, and power generation using reactive metals, zinc, aluminum, or elemental carbon with air-depolarized cathodes. He led the DOE National Program to develop novel metal-air fuel batteries for electric vehicle propulsion. He assembled and led a team to develop advanced processes for the growth and production of optical crystals for lasers. In addition, he has led projects to develop advanced processes, such as molten-salt oxidation, for treating mixed waste and military waste.

Currently, Cooper is scientific capability leader for electrochemistry and corrosion in the Chemistry and Materials Science Directorate. He is also the technical director of a private-sector collaboration to develop zinc-air fuel cells and batteries and is the inventor-director of projects to develop practical high-efficiency carbon-oxygen cells for mitigating the greenhouse gas emissions associated with electric power generation.